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INFLUENCE OF COMPOSITION AND PROCESS SELECTION ON DENSIFICATION OF SILICON NITRIDE

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CERAMICS RESEARCH DIVISION

May 1982

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ABSTRACT

In the development of fully dense silicon nitride, both compositional selection and microstructural control are taken into consideration to maximize resultant properites. The effect of impurities and phase behavior on high temperature properties depends on the silicon nitride-additive compositional selection. Both strength and fracture toughness within a given compositional system can be improved by process control of the microstructure, i.e., grain size and morphology. Dual pressure processing of silicon nitride-yttria-alumina materials can be used to sinter these materials to near theoretical density.

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1 INTRODUCTION

Until approximately five to six years ago, dense Si_3N_4 was primarily produced by hot pressing with the use of an appropriate densifying additive. Recently, due to economic advantages and the necessity to produce complex shapes, sintering and hot isostatic gas pressing have received increasing attention. Adopted modifications of the sintering approach include the use of N_2 gas overpressure to 10 MPa to suppress thermal decomposition of the Si_3N_4, and the development of a dual N_2 gas pressure procedure where densification from the closed pore stage proceeds under a higher N_2 gas pressure than used for the initial sintering step. The sintering approach has also been facilitated by the development of improved Si_3N_4 starting powders, i.e., having higher surface area and greater uniformity. The selection of the additive composition for densification is still primarily based on prior successful hot pressing results. Initial screening and evaluation of new compositional systems is quickly performed by hot pressing. It has been demonstrated that Si_3N_4 is adaptable to compositional and/or microstructural alterations which can improve the properties and performance of the material. The primary material parameters considered for alteration are shown in Table 1. The compositional approach involves additive selection, impurity effects, and phase equilibria/behavior studies. Microstructural improvement relies on starting material characteristics and optimum selection of processing parameters.

Most studies concerned with the densification of Si_3N_4 have concentrated on the use of MgO, Al_2O_3, or Y_2O_3 as densification aids. More recently, CeO_2 and ZrO_2 have received attention. The criteria for additive selection generally emphasized resistance to high temperature creep and oxidation. Often, the selection of additive composition and quantity was problematic in that promoting ease of densification of Si_3N_4 bodies resulted in a degradation of their high temperature properties.

Table 1. Material Parameter

Goal	Approach	
	Compositional Control (Grain Boundary Eng.)	Microstructural Control
Improved Performance Reliability of Si_3N_4 by Process Control	Additives (MgO, Y_2O_3, etc.)	Grain Size
	Impurities (Metal Cations, Carbon)	Grain Morphology (Equiaxed/Prismatic)
	Phase Behavior (Multicomponent Systems)	Phase Control (α/β ratio)

2 MICROSTRUCTURAL EFFECTS

Studies on the use of MgO additive have been well documented in the technical literature. Within the past few years, these studies have been increasingly concerned with microstructural development and its influence on resultant properties of Si_3N_4. Since the early observation that high alpha phase starting powders were required to produce the highest strength Si_3N_4 product, the effect of microstructure and microstructural modifications produced by changes in composition and process parameters were generally neglected until Lange (1) reported on the morphological development of beta grains in hot-pressed Si_3N_4. Knoch and Gazza (2) subsequently investigated the influence of Si_3N_4 starting powders with different alpha/beta phase content on the modulus of rupture of hot-pressed Si_3N_4-5%MgO composition using different time, temperature, and pressure parameters. In this study, the most dramatic effect of process parameter variation on the resultant microstructure and properties was produced by hot pressing Si_3N_4-5%MgO for various times from 10 minutes to three hours at 1600°C using 70 MPa uniaxial pressure. The room temperature modulus of rupture values and resulting β-fraction associated with these hot-pressing parameters are shown in Figure 1. Maximum strength is reached at nearly full conversion of α phase to β phase with interlocking elongated grain morphology. Further coarsening of the grain size by extending the hot-pressing time produces a reduction in strength.

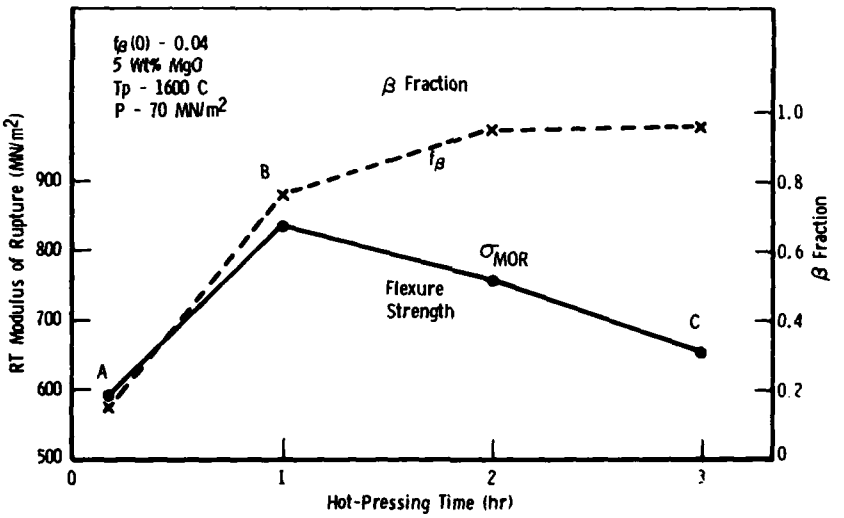

Figure 1. Effect of hot-pressing time on flexural strength and β-fraction of hot-pressed Si_3N_4-5%MgO.

1. LANGE, F. F. *Fracture Toughness of Si_3N_4 as a Function of Initial α-Phase Content.* Technical Report No. 4, Office of Naval Research, Contract N00014-77-C-0441, July 1978.
2. KNOCH, H., and GAZZA, G. E. *On the α to β Phase Transformation and Grain Growth During Hot Pressing of Si_3N_4 Containing M_gO.* Ceramurgia International, v. 6, no. 2, 1980, p. 51-56.

Fracture toughness (K_c) of these samples, shown in Figure 2, were determined by the indentation method (3). The fracture toughness increased to a maximum of approximately 7 $MNm^{-3/2}$ with increasing α to β transformation and subsequently dropped to 4.5 $MNm^{-3/2}$ as the grain size coarsened and the aspect ratio decreased. Fracture toughness results from other studies (4,5) are also shown in Figure 2. Grain coarsening effects on K_c appear to be similar to those observed by Himsolt, et al (5).

3 PHASE BEHAVIOR AND IMPURITY EFFECTS

The development of more refractory Si_3N_4 materials using Y_2O_3, CeO_2, etc., tended to concentrate more on compositional alterations and phase relations. In particular, Y_2O_3 additions were found to form a higher viscosity glassy phase and quaternary oxynitrides. Although the properties of the Y_2O_3-doped Si_3N_4 were found to be excellent, particularly at temperatures above 1200°C, certain intermediate temperature phase instability was found at temperatures between 700°C and 1100°C in an oxidizing environment (6). The instability was attributed to large changes in the molar volume of the secondary phase and its oxidation product. However, the phase instability and resultant crack formation was not always found in oxidized Si_3N_4 bodies containing these quaternary phases.

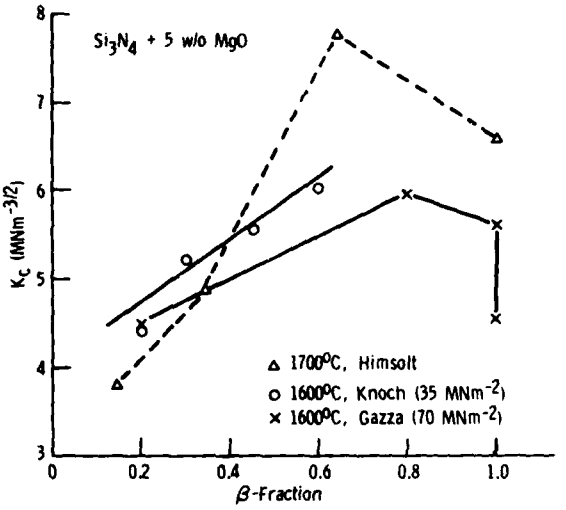

Figure 2. Variation of fraction toughness with β-fraction in hot-pressed Si_3N_4-5%MgO.

3. EVANS, A. G., and CHARLES, E. A. *Fracture Toughness Determinations by Indentation.* Journal of American Ceramic Society, v. 59, 1976, p. 371-372.
4. KNOCH, H., GAZZA, G. E., and KATZ, R. N. *The Influence of Processing Parameters on Development of Microstructure in Hot-Pressed Silicon Nitride and Its Correlation With Mechanical Properties.* Proceedings of the 4th CIMTEC Meeting, Saint Vincent, Italy, 1979, p. 737-750.
5. HIMSOLT, G., HUEBNER, H., KLEINLEIN, W., and KNOCH, H. *Mechanical Properties of Hot-Pressed Silicon Nitride With Different Grain Structures.* Journal of American Ceramic Society, v. 62, no. 1-2, 1979, p. 29-32.
6. LANGE, F. F., SINGHAL, S. C., KUZNICKI, R. C. *Phase Relations and Stability Studies in the Si_3N_4 - SiO_2 - Y_2O_3 Pseudoternary System.* Journal of American Ceramic Society, v. 60, no. 5-6, 1977, p. 249.

In related studies, Knoch and Gazza (7) at AMMRC and Schoun (8) at NASA found impurity influences associated with this behavior. In the former study, the presence of carbon or silicon carbide in hot-pressed Si_3N_4-Y_2O_3 (melilite) specimens caused extensive cracking during oxidation at temperatures of 800°C to 1000°C while carbon-free material exhibited parabolic oxidation kinetics. When a carbon-containing Si_3N_4 powder with a carbon content of approximately 0.6% was used to form the melilite compound, degradation occurred during oxidation. Subsequently, it has been found that a carbon content as low as 0.2 to 0.3% in Si_3N_4 containing melilite phase will crack during oxidation. The extent of degradation may also depend on the amount and distribution of melilite phase in Si_3N_4. Schoun (8) demonstrated that the presence of tungsten in sintered Si_3N_4 containing the melilite phase produced severe oxidation and cracking at 750°C while similar Si_3N_4-8%Y_2O_3 material containing 2%WC or no intentional impurity showed only small weight gain.

Whether severe oxidation and cracking of Si_3N_4-Y_2O_3 compositions containing melilite phase, K-phase, or J-phase is observed or not, most studies indicate that the oxidation rates of Si_3N_4-Y_2O_3 compositions within these phase fields are higher than for compositions within the Si_3N_4-Si_2N_2O-$Y_2Si_2O_7$ triangle (6,9) or Si_3N_4-$Y_2Si_2O_7$-$Y_5Si_3O_{12}N$ triangle (10). Optimum Si_3N_4-Y_2O_3 compositions with regard to the best combination of strength, oxidation, and creep resistance should be found within the latter two compatibility triangles.

4 COMPOSITIONAL MODIFICATIONS

The successful use of Y_2O_3 and CeO_2 as densification aids to Si_3N_4 suggested that other rare earth oxides might also be effective additives. Andersson and Bratton (11) studied the rare earth oxides of Y, La, Ce, Pr, Nd, Sm, Gd, Dy, Er, and Yb with compositions within the compatibility triangle Si_3N_4-Si_2N_2O-$M_2Si_2O_7$. Additionally, they included Sc_2O_3, NiO, Cr_2O_3, and ZrO_2 in their study. The highest silicate composition ($M_2Si_2O_7$ for most additives studied) was chosen to maintain stability with respect to SiO_2 and retain good oxidation resistance. Incipient melting points were determined for Si_3N_4-M_xO_y-SiO_2 compositions as well as elevated temperature flexural strengths. Both parameters are shown plotted in Figure 3 as functions of the ionic radii of the rare earth elements used for each composition.

7. KNOCH, H., and GAZZA, G. E. *Effect of Carbon Impurity on the Thermal Degradation of an Si_3N_4 - Y_2O_3 Ceramic.* Journal of American Ceramic Society, v. 62, no. 11-12, 1979, p. 634-635.
8. SCHUON, S. *Effect of W and WC on the Oxidation Resistance of Yttria Doped Silicon Nitride.* NASA TM-81528, Presentation at American Ceramic Society Meeting, Chicago, Illinois, 28-30 April 1980.
9. RAE, A. W. J. M., THOMPSON, D. P., and JACK, K. H. *The Role of Additives in the Densification of Nitrogen Ceramics.* Ceramics For High Performance Applications-II. Proceedings of the 5th Army Materials Technology Conference, Newport, Rhode Island, March 1977.
10. QUACKENBUSH, C. L., and SMITH, J. T. *Phase Effects in Si_3N_4 Containing Y_2O_3 or CeO_2: II, Oxidation.* American Ceramic Society Bulletin, v. 59, no. 5, 1980, p. 533-537.
11. ANDERSSON, C. A., and BRATTON, R. *Ceramic Materials For High Temperature Turbines.* Final Technical Report, U.S. Energy Res. Dev. Adm. Contract EY-76-C-05-5210, August 1977.

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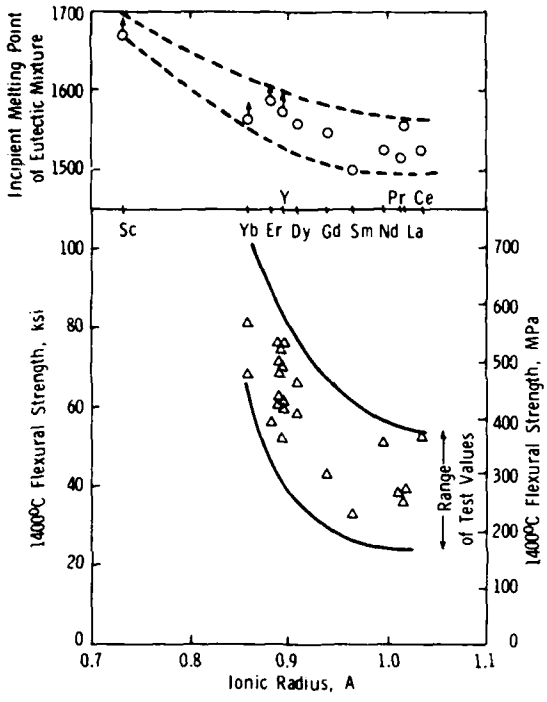


Figure 3. Effect on ionic radius of rare earth elements on the eutectic melting points and elevated temperature strengths of Si_3N_4-M_xO_y-SiO_2. (Ref. 11)

The ionic radii were selected because they are indicative of the bond strengths of those rare earths in the $M_2Si_2O_7$ structures. Both the elevated temperature flexural strength and eutectic melting points of the Si_3N_4-Si_2N_2O-$M_2Si_2O_7$ systems increase with decreasing rare earth ionic radii, i.e., with increasing bond strength. The high temperature property potential of Sc-containing Si_3N_4 is suggested on the plot and recent work by Morgan, Lange, et al (12) illustrates the excellent properties achieved with the Si_3N_4-Sc_2O_3-SiO_2 system. The cost of Sc_2O_3 may limit its usefulness, however, beyond the research stage. Further property improvement was suggested by alloying various rare earth pyrosilicates or by substituting smaller ions, e.g., Al^{+3}, Cr^{+3}, within solubility limits in the monoclinic structures.

As processing emphasis of dense Si_3N_4 gradually shifted from hot pressing to sintering, accommodations in additive selection were necessary to insure a sufficient amount of liquid phase formed to promote densification. Additionally, the liquid had to form at temperatures where the system was thermally stable, and be sufficiently reactive with the Si_3N_4 to produce densification by the mechanism of solution-reprecipitation. The addition of Al_2O_3 to the Si_3N_4-Y_2O_3 system is effective in promoting sinterability over Y_2O_3 additions alone. Excellent RT strength values are obtained using the combined Y_2O_3-Al_2O_3 additions to Si_3N_4 but high temperature properties can be adversely affected by the formation of low viscosity Y-Si-Al-O-N glasses. The high temperature property studies of Smith and Quackenbush (13) have shown that for

12. MORGAN, P. E. D., LANGE, F. F., CLARKE, D. R., and DAVIS, B. I. Journal of American Ceramic Society, Communications Section, v. 64, no. 4, 1981, p. c77.
13. SMITH, J. T., and QUACKENBUSH, C. L. *Phase Effects in Si_3N_4 Containing Y_2O_3 or CeO_2: I. Strength.* American Ceramic Society Bulletin, v. 59, no. 5, 1980, p.529-532,537.

Si_3N_4 containing 4 to 13% Y_2O_3, the Al_2O_3 addition should be less than 2% to approach high temperature property requirements of gas turbine engine components. For maximum high temperature creep and oxidation resistance, the Al_2O_3 should be minimized or eliminated. The effect of small Al_2O_3 additions on the stress-rupture life of a commercially pure Si_3N_4-7.5%Y_2O_3 composition is shown in Figure 4. A stress-rupture temperature of 1300°C and a step temperature-stress rupture (STSR) cycle of 1000°C to 1300°C was used with applied stresses of 140 MPa (20 ksi) and 200 MPa (30 ksi). As shown in the figure, a reduction in the amount of added Al_2O_3 from 2% to 0.5% dramatically increases the time to failure under the given stresses. Reducing the stress level from 200 MPa (30 ksi) to 140 MPa (20 ksi) produces a $2\frac{1}{2}$ to 3 times increase in stress-rupture life. The slight difference in time to failure between the stress-rupture and STSR values may be due to a small contribution to slow crack growth at temperatures below 1300°C. In the STSR test, the specimen were held under stress at temperatures of 1000°C, 1100°C, and 1200°C for 24 hours at each temperature before reaching the final test temperature of 1300°C. As a comparative point of reference, STSR conditions (1300°C, 200 MPa) resulted in an approximately 15-hour failure life for hot-pressed NC-132 (14). The use of higher purity Si_3N_4 starting material rather than the commercially pure grade should shift the curves shown in Figure 4 toward longer time to failure.

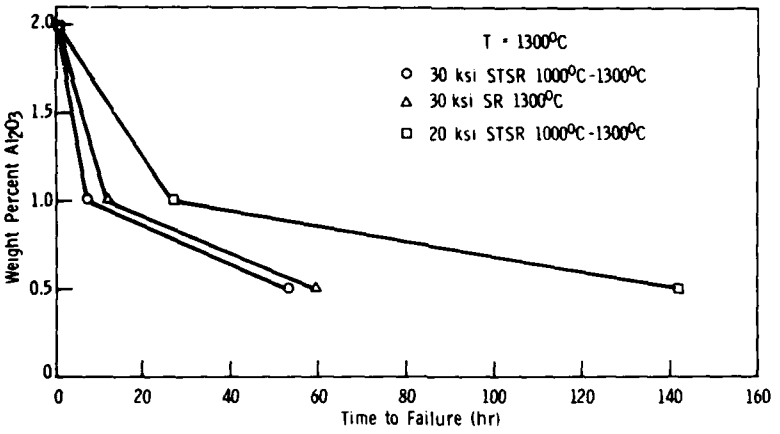


Figure 4. Effect of Al_2O_3 additions on the stress-rupture life of a Si_3N_4-7.5%Y_2O_3 base composition.

5 GAS PRESSURE SINTERING

As processing emphasis shifts from hot pressing to sintering, modification to conventional sintering procedures must be explored to overcome densification problems associated with low diffusivity

14. QUINN, G. D., and QUINN J. B. *Slow Crack Growth in Hot-Pressed Silicon Nitride.* Proceedings of The International Symposium on the Fracture Mechanics of Ceramics, Penn State University, Pennsylvania, July 1981.

and dissociation. The use of high N_2 gas pressure for sintering was found to be effective in promoting densification while limiting decomposition of the material (15-17). The need to densify complex shapes of Si_3N_4 requires that isostatic gas pressure be used. Conventional hot isostatic pressing where pressures greater than 70 MPa are normally used has been successful in producing Si_3N_4-5%Y_2O_3 material with excellent high temperature properties (18). A cladless dual N_2 pressure technique has also shown good results in producing near fully dense ceramics (19). Greskovich and Palm (20) reported using the two-step method to produce 99+% dense $Si_3N_4/SiBeN_2$ composition. Recently, Gazza et al (21) densified $Si_3N_4/Y_2O_3/Al_2O_3$ compositions using 0.1 MPa N_2 pressure for initial sintering, then raising the pressure to 2.0 MPa to produce final densification.

The compositions used in this study are shown in Figure 5, a plot of density versus N_2 pressure. Some additive compositions were prereacted to form compounds, e.g., $Y_3Al_5O_{12}$, $Y_4Al_2O_9$, before mixing with the Si_3N_4 powders. Cold-pressed specimens were initially sintered under 0.1 MPa N_2 pressure. Some compositions sintered to the closed pore stage. In the second sintering step, the pressure was raised to 2.0 MPa while the temperature was held at 1770°C to 1780°C. Ninety-minute hold times were used for each step. As shown in Figure 5, all compositions exhibited increased densification. Those which were initially sintered to the closed pore stage were densified to near maximum density. The poor initial sintering behavior of the 117 and 113 compositions is probably related to lack of sufficient liquid formed with the available silica in the system.

Differences in resultant properties and microstructures were observed which were influenced by the starting Si_3N_4 powder. In Figure 6, microstructures obtained from replicas on polished and etched surfaces and scanning electron micrographs on fracture surfaces are shown for dual-pressure sintered Si_3N_4-10%YAG compositions. Finer grain size and higher grain aspect ratios appear to be developed in the LC-10 powder for the processing conditions used. Room temperature modulus of rupture measurements on both 112 and 105 materials reflect the microstructural differences. An average MOR of 779 MPa (113 ksi) was observed for the finer grain specimen while the coarser 105 specimen had an average MOR of 538 MPa (78 ksi).

15. MITOMO, M., TSUTSUMI, BANNAI, E., and TANAKA, T. *Strengthening of Si_3N_4.* American Ceramic Society Bulletin, v. 55, no. 3, 1976, p. 313.
16. PRIEST, H. F., PRIEST, G. L., and GAZZA, G. E. *Centering of Si_3N_4 Under High Nitrogen Pressure.* Journal of American Ceramic Society, v. 60, no. 1-2, 1977, p. 81.
17. MITOMO, M. *Pressure Sintering of Si_3N_4.* Journal of Materials Science, v. 11, no. 6, 1976, p. 1103.
18. WILLS, R. R., BROCKWAY, M. C., McCOY, L. G., and NIESZ, D. E. *Preliminary Observations on the Hot Isostatic Pressing of Silicon Nitride.* Proceedings Ceramic Engineering and Science, v. 1, no. 7-8, 1980, p. 534-539.
19. HARDTL, K. H. *Gas Isostatic Hot Pressing Without Molds.* American Ceramic Society Bulletin, v. 54, no. 2, 1975, p. 201-205, 207.
20. GRESKOVICH, C., and PALM, J. *Development of High Performance Sintered Si_3N_4.* Final Technical Report SRD-80-111, September 1980, Contract No. DAAG-46-78-C-0058, under AMMRC/DOE Interagency Agreement EC-76-A-1017-002.
21. GAZZA, G. E., and KATZ, R. N. *Development of Sinterable Si_3N_4.* Army Materials and Mechanics Research Center, AMMRC SP 80-5, November 1980; also presented at the Department of Energy Automotive Technology Development Contractor's Coordination Meeting, Dearborn, Michigan, November 1980.

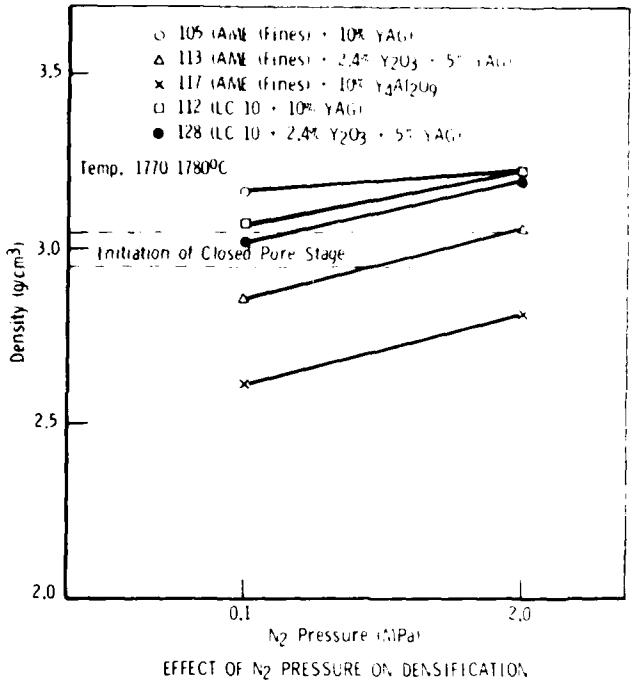
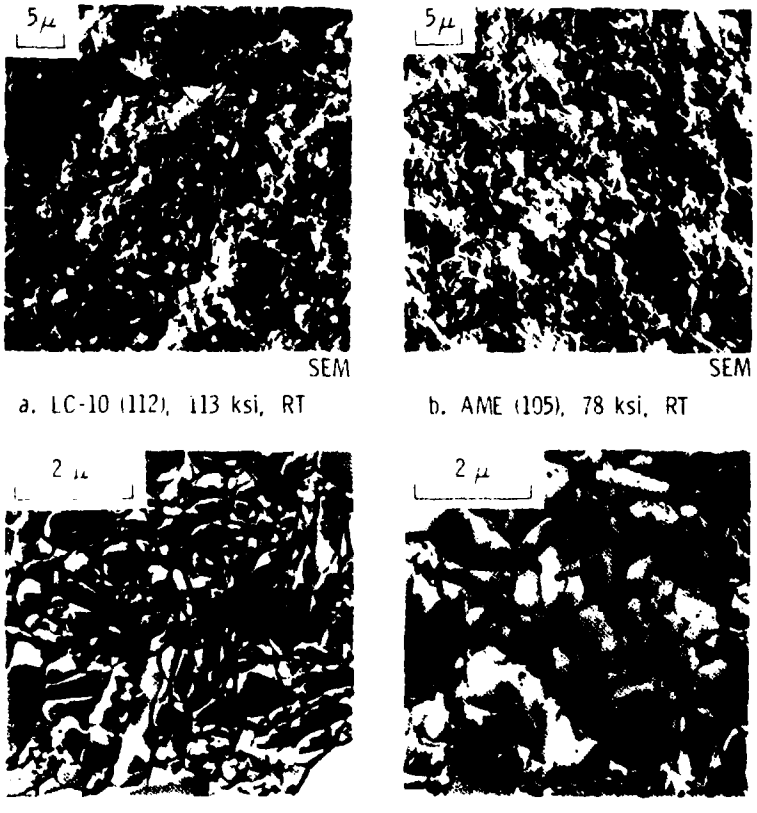


Figure 5. Effect of Dual N_2 pressure process on densification on various Si_3N_4-Y_2O_3-Al_2O_3 compositions.



a. LC-10 (112), 113 ksi, RT

b. AME (105), 78 ksi, RT

Figure 6. Microstructures of sintered Si_3N_4-10%YAG; fractographs (upper) and surface replicas (lower).

6 SUMMARY AND CONCLUSIONS

Studies on the hot-pressing of various Si_3N_4/metal oxide systems demonstrate that both compositional and microstructural alterations can be used to produce significant property differences within a given materials system. For Si_3N_4/MgO, microstructural studies demonstrate the need for judicious selection of starting materials and a more exact determination of optimum process parameters required for densification. For the Si_3N_4/Y_2O_3 materials system, it appears that composition restricted to the Si_3N_4-Si_2N_2O-$Y_2Si_2O_7$ or Si_3N_4-$Y_2Si_2O_7$-$Y_5Si_3O_{12}N$ phase fields will have the best combination of high temperature properties. The role of impurities within this compositional system needs further elucidation.

The addition of Al_2O_3 to the Si_3N_4/Y_2O_3 system facilitates densification but appears to degrade high temperature properties with some dependence on the amount of Al_2O_3 added. This paper did not consider grain boundary crystallization techniques which may alleviate the formation of low viscosity phases.

Emphasis on densifying high purity Si_3N_4/Y_2O_3 compositions appears to be emerging in the area of hot isostatic gas pressing. Techniques requiring cladding have been successful in producing dense, high strength bodies. Cladless hot isostatic gas pressure methods using N_2 pressures less than 10 MPa have also been effective in producing dense bodies with strength and microstructure similar to hot-pressed material.

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